

7. RADIOACTIVITY AND TOXICITY OF COOLANTS

7.1. SODIUM COOLANT RADIOACTIVITY

“Operational” radioactivity. The natural isotope of sodium is ^{23}Na (abundance 100%). Neutron capture processes in sodium (n,γ reaction resulting in 1.4 MeV γ -quanta emittance) lead to formation of ^{24}Na isotope with half-life time of 15 hours, while sodium is flowing through the core. Besides, there is $(n, 2n)$ threshold reaction producing ^{22}Na with 2.6 years half-life time. ^{22}Na emits 1.3 MeV γ -quanta; its activity is proportional to thermal power of the reactor plant.

Neutrons are not practically generated by sodium radionuclides with half-life time exceeding 2.6 years value, even after 50 years of exposition to intensive fast neutron flux.

^{24}Na is the main isotope giving rise to requirement of protection against γ -radiation. Approximately 10 days after reactor shutdown the primary circuit activity is mainly determined by ^{22}Na .

This feature, along with the fact that sodium interacts chemically with water and air, results in three circuit reactor power plant design, including:

- primary circuit containing radioactive sodium heated up in the core,
- secondary non-radioactive sodium circuit coupled with the primary circuit by the intermediate heat exchanger,
- tertiary water circuit producing steam for electricity generation.

Within the primary circuit radioactive coolant is protected against air by the steel barriers and cover argon gas. Radioactive sodium of the primary circuit is separated from non-radioactive sodium of the secondary circuit by the steel tubes of the intermediate heat exchangers.

The operating experience of different reactors has shown, that the coolants separated by the wall of heat exchanging component of the reactor heat removal circuit (steam generator in PWR, turbine condenser in BWR and intermediate heat exchanger in LMFR) should be the same to assure nuclear plant safety and reliability (or at least highly compatible to each other in order to avoid plugging of the core flow channels by the products of coolants interaction). The rise of the total NPP cost owing to this measure (application of identical coolants in the primary and the secondary circuits) and use of tertiary water-steam circuit is insignificant, since standard operation and maintenance of steam generator, water circuit and turbine plant are applied.

Operational experience gained on LMFRs, such as BN-350, Phenix, PFR, BN-600 etc. has proved, that sodium is practically non-corrosive to stainless steel, and content of impurities, mainly oxygen and carbon, are maintained at acceptably low level by the cold traps installed in the bypass of the main coolant circuit. Radioactive hazardous isotopes (caesium, tritium, strontium and iodine) are retained by sodium.

The primary sodium activity in the reactor under operation, mainly determined by ^{24}Na (BN-350 reactor, Kazakhstan) is about 10 Ci per kilogram of sodium [7.1], while after the reactor has been shut down for decommissioning residual activity of ^{22}Na is $\sim 1 \times 10^{-4}$ Ci per kilogram of sodium [7.2].

If any component of the primary sodium circuit is to be removed from the reactor for the purpose of repair/maintenance, sodium sticking to the surface must be removed because of (a) chemical reaction of sodium with oxygen and moisture in air, and (b) radioactivity of sodium. The process of radioactive sodium removal has been selected for the reasons of safety, effective cleaning of different components, economics, etc.

Cover gas is another source of LMFR radioactivity. Primary gas activity is to considerable extent determined by the impurities in sodium and activation of ^{40}Ar and ^{41}Ar . As a result of (n, p) reaction, radioactive ^{23}Ne with short half-life of 38 s is produced from ^{23}Na [7.3].

Residual radioactivity. As it was pointed out in [7.4], the amount of the long lived radioactivity generated in sodium by neutrons is negligible. Activation of sodium reaches equilibrium state in about ten years of the first cycle of its use and will never exceed this level. The long-lived radionuclides furnished by fission products, sodium impurities and corrosion activation products are chemical elements alien to sodium, that makes possible its external contamination at the reactor plant decommissioning stage.

Two strategies have been considered for the disposal of contaminated sodium:

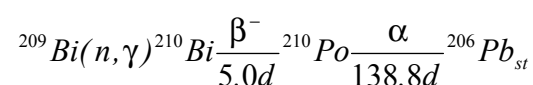
- separation of radioactive products from sodium, reuse of the purified sodium and conversion of the concentrated waste into an inert form for permanent repository,
- retaining of radioactive sodium on the plant site for later use.

Analysis has shown [7.4], that sodium coolant of 1 GW(e) LMFR after 50 years of operation and ~ 50 years of retaining may be exempted for free use in the industry or returned to the nature.

It was pointed out at the IAEA consultants meeting on the BN-350 reactor (Kazakhstan) decommissioning (Obninsk, Russian Federation, 23–27 Feb. 1998), that the main radionuclides determining sodium radioactivity are Na-22 ($T_{1/2}=2.6$ a), Co-60 ($T_{1/2}=5.3$ a) and Cs-137 ($T_{1/2}=30$ y).

7.2. LEAD-BISMUTH AND LEAD COOLANTS RADIOACTIVITY

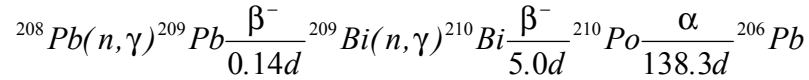
“Operational” radioactivity. The natural isotope of bismuth is Bi-209 (abundance $\sim 100\%$). Neutron capture results in the formation of Po-210 by the following reaction:



The small amount of Po-209 is formed from Po-210 by (n, 2n) reaction.

Po-210 having 138 days half-life undergoes α -decay, and Po-209 decays in a similar way with a half-life of 120 years. Polonium is volatile at coolant operating temperatures, some amount of it migrating to the cover gas, where it forms aerosols [7.6, 7.6a]. Leaks of cover gas or coolant may cause contamination problems and maintenance of components requires special measures to protect personnel.

Even in case of lead coolant the problem of polonium contamination exists because of Bi-209 formation by neutron capture in Pb-208 (abundance 52.3%) [7.5]:



Although the rate of production of Po-210 by this two-step process is much lower (1000 times lower), fraction of polonium migration out of the coolant is higher (perhaps, by ~100 times) because of higher coolant temperature.

In the report [7.5] by V. Orlov, et al. it is pointed out, that in the reactor cooled by lead-bismuth polonium activity value is determined by reaction on ${}^{209}\text{Bi}$ isotope, and equilibrium activity is ~10 Ci/kg. As for lead cooled reactor ($5\cdot 10^{-4}\%$ Bi) polonium activity is determined by reaction on both ${}^{209}\text{Bi}$ and ${}^{208}\text{Pb}$ isotopes and may reach $5\cdot 10^{-4}$ Ci/kg value by the end of the reactor service life.

Authors of the report [7.5] have noted, that such lead-bismuth coolant activity gives rise to the problems even under normal operating conditions. They consider that in case of the cover gas leak rate of 0.01% of its volume per day, release of ${}^{210}\text{Po}$ to the central hall may cause (if gas circuit is not cleaned from polonium) 200 fold exceeding of its maximum permissible concentration (mpc) [7.1]. To ensure that mpc value is not exceeded for personnel in the central hall, it is necessary to comply with very strong requirements for the reactor cover gas circuit leak-tightness.

Residual radioactivity. Experts in nuclear technology believe that selection of coolant for the future wide-scale nuclear facilities with fast neutron spectrum should be done taking into account management and disposal of the radioactive waste arising from NPP decommissioning. Specialists suppose that post operational characteristics of a reactor plant including spent coolant waste should be considered as highly important as those related to the operation stage. The primary circuit coolant of the NPP could be one of the main potential sources of radioactive waste.

Long term residual activity of sodium, lead-bismuth and lead coolants of fast reactors have been studied and compared by V. Ousanov, D. Pankratov, et al. [7.4]. It was found, that specific α -activity of typical lead-bismuth coolant is determined by Bi-210m (half-life = 3.6×10^6 years) generated in reaction Bi-209 (n, γ) Bi-210m. The long lived β -activity of Bi-208 (half-life = 3.65×10^5 years) is produced in the reaction Bi-209 (n, 2n) Bi-208.

The most important contributor to specific long-lived residual radioactivity of lead coolant is Pb-205 (half-life = 1.51×10^7 years) generated in the reaction Pb-204 (n, γ) Pb-205. The specific β -activity of the pure lead coolant is significantly lower than that of lead-bismuth. Activation of lead-bismuth and lead coolants will increase in every stage of recycling, if recycling is possible in principle [7.4].

Thus, the residual activity of lead-bismuth and lead coolants is expected to be as high as millions of years. As it is pointed out in [7.4], purification of lead-bismuth and lead coolants from the long lived radionuclides of bismuth and lead (if it is possible) would be too expensive.

In the summary [7.4] it was concluded that “the sodium coolant appears to be the most appropriate for realisation of the non-waste goal among three liquid metal coolants considered”.

Lead-bismuth purification from polonium. Some studies in this field have been carried out at the IPPE (Obninsk, Russian Federation). In particular, the possibility of polonium alkaline extraction from lead-bismuth was studied, coolant flow rate of ~0.1% being sufficient to decrease polonium activity in the circuit by over 4 orders and transform extracted polonium into non-volatile compounds. Periodic polonium removal from the circuit is possible for instance before scheduled repair.

It is important to assess evaporation rate of the media, where Po is present. Po can be either in elementary form or in chemical compounds, such as oxide (PoO), inter-metalloid (PbPo), hydride (PoH), etc. Evaporation rate depends on the type of composition, its temperature, density, gas content and other parameters. Rate of evaporation into vacuum follows the Lenthmure law:

$$G_o = kP\sqrt{M/T} , \quad (7.1)$$

where: P-pressure, K = 0.0044, M — molecular mass, T — temperature. If the substance is in solution, evaporation rate is proportional to mole part “m”: $G=mG_o$.

Feuerstein experiments [7.7, 7.8] on Po evaporation from eutectic Pb-Li-17 have shown that within 300–800°C temperature range, evaporation rate of Po into vacuum is 10^3 times lower than that of elementary Po. Inert gas atmosphere (Ar, Ne) reduces evaporation rate by 10^3 times more. Similar data was obtained by Tupper [7.9] in the experiments on Po evaporation from PbBi.

Thus, Po may be thought of as evaporated from PbBi in the form of PbPo or BiPo. Saturation pressure for polonide vapour follows the relationship:

$$\lg P=6.94-(7270/T). \quad (7.2)$$

Calculations made using the relationships presented give the following polonium yield rates:

From PbBi with activity of $1.85 \cdot 10^{13}$ Bq/L at 300°C	-	$6.7 \cdot 10^5$ Bq/kg
From Pb with activity of $1.85 \cdot 10^{10}$ Bq/L at 500°C	-	$1.15 \cdot 10^6$ Bq/kg

Although Po activity in lead is 1000 times lower than that in PbBi, polonium yield from lead is approximately the same due to higher temperature.

Polonium and reactor engineering. In the overviews by B. Gromov, D. Pankratov, et al [7.9, 7.10] general conclusion was made on that Po-210 activity cannot be considered as the main barrier preventing use of PbBi coolant. This conclusion is based to considerable extent on the operating experience of NPP, nuclear submarines and their ground-based prototypes. Results of studies on beyond design accidents on PbBi cooled stationary reactors are presented in [7.11].

Experience gained on LOCA studies has shown that application of protective coating makes it possible to confine polonium by preventing its release to the environment. This coating effect is based on sorption and dissolution of impurities in dispersed medium and fixing in the coating.

In case of steam generator tube failure and depressurization of the secondary circuit, PbBi alloy can find its way into the secondary circuit and water can be contaminated by polonium. In this case basic amount of polonium is kept in the alloy, condensate saturation activity of $10 \div 10^4$ Bq/kg is reached, and SG inner surfaces become contaminated owing to polonium sorption from the water. Water evaporation determines radioactivity level in the turbine hall.

Contamination of the inner surface of the secondary circuit gives rise to danger in case of equipment repair, since the alloy kept in the secondary circuit is a permanent source of water contamination. Water replacement and SG inner surface decontamination without any alloy removal would not give the desired result. The secondary circuit decontamination turns out to be possible only after complete removal of the alloy from the secondary circuit. Obviously, this cannot be assured.

If SG tubes failures are assumed possible, the alloy penetration to the secondary circuit should not be neglected. Tupper's experiments [7.9] have shown that PbBi evaporation rate is within the limits given by Raoult law. Lead contained in air may produce the same danger for health as ^{210}Po does.

Polonium activity is one of the important operational problems in case of lead-bismuth used as a coolant. To decrease this problem lead has been proposed instead of lead-bismuth. This has the following consequences:

1. Such problems as control and maintenance of coolant quality, its compatibility with structural materials, corrosion strength, etc. should be studied to a great extent at higher temperatures. Though lead is similar to some extent to lead-bismuth, nevertheless it is a different coolant.
2. Some engineering problems are caused by thermal insulation and heating of loops up to the temperatures over 400°C . Related system should have high reliability, assuring required conditions in both operation and repair stages.
3. There is a problem of elimination of coolant freezing under any accident conditions (failure of heaters, quick pressure decrease in the water circuit, etc.), or its subsequent melting without structure damage.

4. Two-circuit heat removal system with steam-water cycle of supercritical parameters is very difficult for realization, because super high pressure steam generator (~250 atm) should be located inside the reactor vessel (BREST concept), and there is a real possibility of the core blockage by the products of water and heavy coolant interaction resulting in the core melt- down. Such incident took place in the propulsion reactor cooled by PbBi [7.11].

The main hazard is α -activity of polonium aerosols present in air and aerosol deposits on the surface. According to the Russian standards, the acceptable specific radioactivity values are specified for personnel: 2.7 Bq/m³ (air) and population: 4.6·10⁻² Bq/m³ (air) and 1 Bq/kg (water and food).

Radioactivity of components. Activity of the primary circuit components is determined by not only natural activity of lead, but also by admixtures (basically Zn, As, Cd and Sb) and corrosion products (See Table 7.1). The activity can be reduced by decreasing Sb content in Pb. The purification technique should be developed.

Among steel corrosion products the greatest contribution to activity is given by Cr-51, Mn-54 and Co-60. LMFR experience shows that corrosion products are deposited on the cold surfaces of the primary circuit. Process of these products deposition from lead is to be studied.

TABLE 7.1. EQUILIBRIUM ACTIVITY OF COMPONENTS

Radionuclides	Half-life time	Equilibrium activity, Bq/L
Zn-65	245 days	2.6·10 ⁸
As-76	26 hours	0.41·10 ¹⁰
Cd-115	54 hours	2.2·10 ⁸
Sb-122	50 hours	0.48·10 ¹⁰
Sb-124	60 days	0.37·10 ¹⁰

Gas activity. The inert gas (Ar) above lead level in the reactor usually contains impurities such as polonium, some amount of hydrogen from oxygen content control systems and tritium. Purification of radioactive cover gas can be made by hydrogen oxidation (to water), water separation and adsorption. Purification from polonium can be made by alkaline extraction. All these methods are considered to develop.

The use of U nitride fuel and boron control rods in LCFR (BREST concept) will cause tritium formation as a result of triple fission on N-14 and B-10 nuclei. Tritium activity may reach 2.6·10⁸ Bq/day, that is rather high value. Because of its penetration into the secondary circuit through SG tube walls, tritium release into the environment is possible. These assessments are thought to be performed not only by calculations but also using experimental studies.

One of the arguments in favour of using lead instead of sodium relates to severe accidents. To justify this argument, an accident causing reactor vessel and building destruction is considered.

The main problem is that of integrity of the fuel elements. Assuming that damaged fuel elements still keep their integrity, and no radioactivity leakage occurs from the fuel, assessments for LMFR and LCFR were made at the RDIPE showing that radioactivity release in LMFR would be 3 to 4 orders higher than that in lead cooled reactor because of sodium burning. However, if the fuel element failures take place (and this seems to be the most probable) coolant radioactivity becomes less important, and fission products would contribute essentially.

7.3. TOXICITY OF LEAD

When in industrial use, lead is released to the atmosphere, and not only its stable isotope, but also radioactive isotopes, such as Pb-210, Pb-212 and Pb-214 are released. Their sources are radioactive gases (including radon).

Lead is highly toxic substance, it is accumulated in the human body and expelled slowly together with the living activity products. Introduced in a human body it attacks the nervous system, marrow, blood and vascular, disturbing albumen synthesis and genetic structure of cells [7.12, 7.13, 7.14].

Maximum permissible concentrations of lead in air and in water are respectively 10^{-5} mg/L and 0.03–0.1 mg/L. In normal reactor operation lead is kept within gas-tight circuit. Enhancement of lead concentration in working rooms is possible as a result of accidents causing leaks in the reactor upper plate, or its disassembly.

TABLE 7.2. HUMAN DISRUPTION INDEX (ENERGY SUPPLY INPUT)

Flow items	Natural baseline flow	Human Disruption Index	Major causes
Lead	25 000 (t/a)	15	Fuel burning associated processes *
Oil to oceans	500 000 (t/a)	10	Oil processing and waste
Cadmium	1 000 (t/a)	8	Associated processes *
SO ₂	50 million (t/a)	1.4	Fuel burning
Methane stock	0.8 ppm	1.1	Agricultural activities
Mercury	25 000 (t/a)	0.7	Associated processes *
Nitrous oxides	10 million (t/a)	0.4	Agricultural activities
Particles	500 million (t/a)	0.25	Fuel burning; land activities
CO ₂	280 ppm	0.25	Fuel burning

* Associated processes include metals processing and manufacturing, but no burning. Abridged from U.N. DEVELOPMENT PROGRAMME (UNDP), Energy after Rio; Prospects and Challenges, UNDP, New York, NY (1997).

Human Disruption Index. General indicator of the impact on the global environment attributed to today's energy activities is Human Disruption Index (HDI) (see Table 7.2). It is essentially ratio of human generated additions to the natural baseline situation for energy related environmental factors such as CO₂, SO₂, NO_x, cadmium, lead, mercury toxic particles, oil to oceans and methane stock. It includes energy chain impacts such as manufacturing that demonstrates the impacts beyond the production stage.

HDI value is 10 for oil put into the oceans, 15 for lead and 0.7 for mercury release corresponding to the human generated movement of 5 million tons of oil put annually into the oceans and also almost 400 000 tons of lead and 20 000 tons of mercury released into the environment. In the context of large natural inventory of radioactive materials in the earth and significant continuous release of natural radon gas to the atmosphere, additions made by nuclear power have a negligible impact on the natural radioactive baseline situation.

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8. COMPARATIVE CHARACTERISTICS OF REACTOR COOLANTS

The comparison of basic characteristics of most of the suitable coolants for fast reactor application was made on the basis of Sections 2–7 and literature data [8.1–8.4] and given below in Table 8.1 The characteristics of steam and gas (helium, CO₂) were included due to their consideration in several countries as a coolants of advanced fast reactors [8.5, 8.6].

It should be pointed out that presently only liquid metal coolant-sodium is widely adopted for fast reactors. Mercury was used for a short period ("Clementine" reactor in the USA and BR-2 in the USSR having, respectively, thermal output powers of 30 and 100 kW). A number of lead cooled fast reactors are being studied presently.

TABLE 8.1 FAST REACTOR COOLANT COMPARISON

Coolant	Advantages	Shortcomings
1	2	3
Sodium	<ul style="list-style-type: none"> • best thermohydraulic characteristics; • low reactor vessel pressure(near atm.); • good breeding characteristics: high BR, short T₂ could be provided; • non-corrosive to stainless steel and any fuel compositions; • decay heat removal by passive means; • low density, preventing fuel SA from floating up, and allowing passive safety rod operation (by gravity force); all the 'dirt' resulted from the assembly and operation is precipitated in the stagnant zones; •retention of caesium, strontium, iodine and tritium hazardous isotopes by sodium; •neutrons do not generate radionuclides with a half life more than 2.6 years (²³Na); 	<ul style="list-style-type: none"> - chemical activity with respect to oxygen and water with explosion potential; - high γ-activity of ²⁴Na; - intermediate circuit necessity; - freezing temperature is about 100⁰C, so heaters are required for maintaining liquid phase of coolant; temperature required for refueling/repairing is ~150–200⁰C; - problems with sodium removal and disposal during reactor plant operation and decommissioning.
Lead, lead-bismuth	<ul style="list-style-type: none"> • lack the high γ-activity; • chemical interaction with water and air without explosion; • low moderation and absorption of neutrons; • good reflecting properties; • good neutronic performance; • reactor decay heat removal by passive means; • high boiling temperature, coolant void reactivity is negative; 	<ul style="list-style-type: none"> - α-radioactive, volatile polonium produced, any leakage from the cover gas poses a hazard to operators and environment(Pb-Bi) - highly corrosive to steels and some fuel compositions; - high density, probability of core pollution by suspensions (assembling and operating 'dirt') in heavy coolants;

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- high freezing temperature (327°C for lead);
 - residual activity of coolant is high ($T_{1/2}$ — $1.5 \cdot 10^7$ y of nuclide ^{205}Pb and $3.6 \cdot 10^6$ y for ^{210}Bi)[8.7];
 - formation of the products of water and heavy metal interaction (SG leaks) which could block flow channels of the core [8.8], two circuit design is questionable;
 - a leak-before-break argument in safety analysis is questionable because of a risk of corrosion damage;
 - problems with radioactive waste management and coolant disposal during decommissioning.

Steam

- possibility of direct cycle or at most two-circuit heat transfer system;
 - transparency;
 - commercial application level;
 - minimum of chemical reactions within the coolant;
 - liquid state at room temperature
 - insignificant activation of coolant itself;
 - less problems with reactor plant decommissioning.
- primary circuit high pressure;
 - serious problem of reactor decay heat removal in the case of loss of forced convection or pipe breaks;
 - high pumping power;
 - high temperature difference between fuel element cladding and coolant;
 - corrosion activity with respect to fuel element structural materials;
 - difficulties maintaining coolant quality to decrease as much as possible its corrosive effect;
 - absence of technological experience of reactor cooling by steam;
 - low breeding ratio;
 - possibility of fission product penetration into a turbine in the case of direct cycle.
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Gas (helium, CO ₂)	<ul style="list-style-type: none"> • two-circuit system (no intermediate circuit); • no activation (helium); • less moderating than sodium and so results in a core with a harder spectrum, which allows good flexibility with regard to plutonium management and minor actinide transmutation; • minimum void reactivity effect; • good compatibility with structural materials; • inertness; • less problems with reactor plant decommissioning; • transparent gas atmosphere facilitates in-service inspection and maintenance • synergy with the technology of HTRs, which already exist or is under development; • principal possibility of direct cycle realization (helium). 	<ul style="list-style-type: none"> - high system pressure; - high pumping power; - necessity for use of fuel element cladding with improved heat transfer surface provision, e.g. by high degree of roughness; - high temperature difference between fuel element cladding and coolant; - serious problem of reactor decay heat removal in the case of loss of forced convection or pipe breaks; - difficulties concerning gas leak from circuit; - absence of commercial scale experience; - low breeding characteristics (for CO₂).
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